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Nuclear polarization and magnetic resonance of unstable ^{170}Tm with beta-ray radiation-detected optical pumping in solids

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Significant nuclear polarization of unstable ^{170}Tm in Tm^{2+}:SrF_2 has been achieved with optical pumping in solids. The hyperfine coupling constant of divalent ^{170}Tm in SrF_2 was thus measured for the first time in high precision with the beta-ray radiation detected magnetic resonance, thereby also determining the nuclear magnetic moment: $0.2476(16)\mu_N$.

Oriented samples of nuclei are of profound utility in solid, nuclear, and particle physics. Among various methods[1] in use of orienting nuclei, optical pumping of atoms in a gas or in a beam is a relatively simple room-temperature method and has been especially used for alkaline-metal and rare-gas atoms.[2] On the other hand, the application of optical pumping in solids for nuclear orientation has not yet been well developed yet although appreciable nuclear polarization has been achieved for some stable nuclei.[3,4] The development of such a technique for orienting, in particular, unstable nuclei in solids is quite desirable for nuclear physics, since the method is rather simple in that it does not require any sub-Kelvin temperature, or high magnetic field, and still it can maintain the appreciable nuclear polarization in solids for a long time after the nuclei are produced in or implanted into the solids.

In this Rapid Communication we present an experimental result on orienting unstable nuclei with optical pumping in solids for the first time and on *directly* detecting their degree of polarization with sensitive detection of optical pumping via the beta-decay asymmetry from polarized nuclei (β-RADOPS; beta-ray radiation-detected optical pumping in solids). The unstable ^{170}Tm in Tm^{2+}:SrF_2 was polarized up to about 4% and the hyperfine coupling constant (hfs) was thus measured in high precision with beta-ray radiation-detected magnetic resonance, thereby determining also the value of the nuclear magnetic moment.

The magnetic circular-dichroism of rare-earth atoms in alkaline-earth fluoride hosts is so large[5] that the electron-spin polarization can be significantly enhanced by pumping with circularly polarized laser light.[3] The electron polarization enhanced by pumping the $4f$-$5d$ band of Tm^{2+} in SrF_2,[6] for example, is then transferred to the nuclei by the hyperfine coupling and/or the selective spin-lattice relaxation processes. The electron polarization can also be transferred to the nuclei by inducing microwave transitions between the relevant hyperfine-Zeeman sublevels (dynamic nuclear polarization[7]). Normally the number of unstable nuclei produced in a crystal via neutron activation or implantation is so small that it is inevitably difficult to detect the nuclear polarization with conventional magnetic resonance (NMR and EPR) methods and even with more sensitive optical methods. The methods of the β-RADOP and/or γ-RADOP thus have to be invoked to get enough sensitivity for the detection of nuclear polarization; the degree of nuclear polarization can be enhanced and/or depressed by applying an rf magnetic field corresponding to a transition between adjacent hyperfine-Zeeman sublevels. Observation of this kind of magnetic resonance with the detection[8] of the β-decay asymmetry resulting from parity nonconservation in the weak interaction is a sensitive method for measuring the hfs constant A, thus enabling us to get the relevant information on the structure of nuclei such as the spin and the magnetic moment.

The overall experimental setup is shown schematically in Fig. 1. A sample of 3×3 mm^2 area and of 1 mm thick crystal of SrF_2 containing 0.02% stable $^{169}Tm^{2+}$ was irradiated by thermal neutrons from the Kyoto University Nuclear Reactor. About one part in 10^6 of ^{169}Tm was converted to unstable ^{170}Tm (half-life is 129 d and nuclear spin I_{170} is 1^-), radioactivity being about 4 μCi. After annealing the sample in vacuum at 250 °C for 10 min, the sample was set in a liquid-helium cryostat in the dc field of an electromagnet. The temperature of the sample was cooled to about 2 K by evacuating the helium chamber. The pumping source was laser light of about 594 nm wavelength from a ring dye laser (Spectra Physics, SP-380-A) pumped by an Ar ion laser (SP-164-09). The direction of the laser light is parallel to that of the applied dc magnetic field. The laser intensity on the whole area of the sample was 150–200 mW. The β rays emitted in the decay of ^{170}Tm were detected with a CaF_2(Eu) scintillator set just behind the sample in liquid helium,

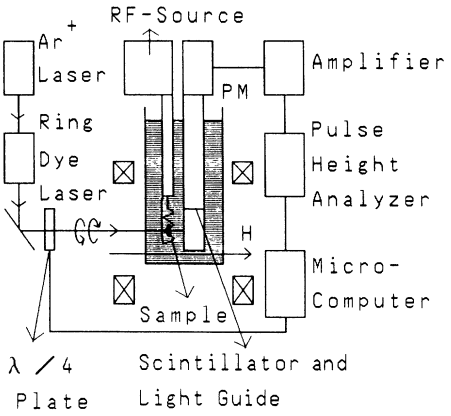

FIG. 1. Experimental arrangement for β-RADOPS, showing optical system, liquid-He cryostat with a sample and β-rays counting system, and data processing system.

and the scintillation light was transmitted via a light guide to a photomultiplier outside the cryostat. After amplification, the signals were fed to and analyzed with a pulse height analyzer. The overall experimental procedure was controlled with a microcomputer.

The sense of circular polarization of the pumping light was reversed every 45 s and the experimental β-decay asymmetry A_{exp} in the β-ray distribution defined by

$$A_{exp}=[N(\sigma^+)-N(\sigma^-)]/[N(\sigma^+)+N(\sigma^-)]$$

was thus measured, where $N(\sigma^i)$ is the number of β rays detected in optical pumping with σ^i polarized light. The value of A_{exp} measured as a function of applied magnetic field with optical pumping is shown in Fig. 2.

The observed A_{exp} was found to oscillate periodically with applied magnetic field. The magnitude of this oscillating variation is shown by the shaded area in Fig. 2.

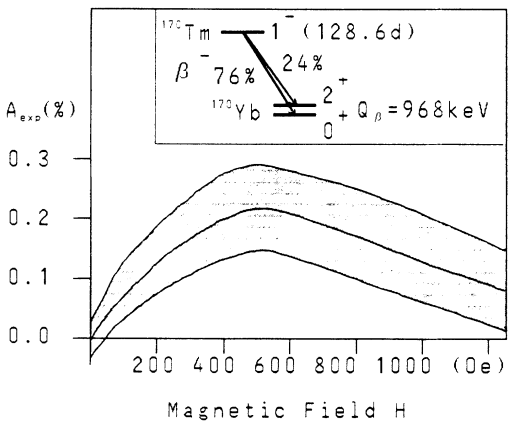

FIG. 2. Experimental β-decay asymmetry A_{exp} observed as a function of applied magnetic field H. The value of A_{exp} oscillates periodically with applied field. The region of this variation is shown within the shaded area, and an averaged value over this oscillation is represented by a solid line. In the inset the decay scheme of ^{170}Tm is shown.

This oscillation is quite reproducible with respect to the applied magnetic field. Detailed discussion of this interesting phenomenon will be given elsewhere.[9] The value of A_{exp} averaged over this oscillation is shown with a solid line in the figure. This mean value of the experimental asymmetry increases with increasing magnetic field up to about 500 Oe and then decreases monotonically. It is estimated that the maximum value of A_{exp} at about 500 Oe corresponds to a nuclear polarization of about 4%.[10] The suppression of the polarization at low field can be qualitatively understood to be due to the effect of cross relaxation between the stable ^{169}Tm and the unstable ^{170}Tm. The decrease of the polarization with increasing magnetic field above 500 Oe, on the other hand, suggests that the nuclear-spin memory effect in the optical pumping cycle[3,11] is smaller at higher fields,[12] where the hyperfine coupling becomes weaker than the electronic Zeeman interaction.

In addition to the optical pumping, a microwave magnetic field of 6.891 GHz was applied to the sample via a five-turn helix coil, and the enhancement of the polarization due to the saturation of the forbidden transition between the substates 2 and 6 [$|M_s=\frac{1}{2}, M_I=0\rangle$ (in the main component) to $|M_s=-\frac{1}{2}, M_I=1\rangle$] [see the energy levels in Fig. 3(c)] was observed at 1.35 kOe. In Fig. 3(a)

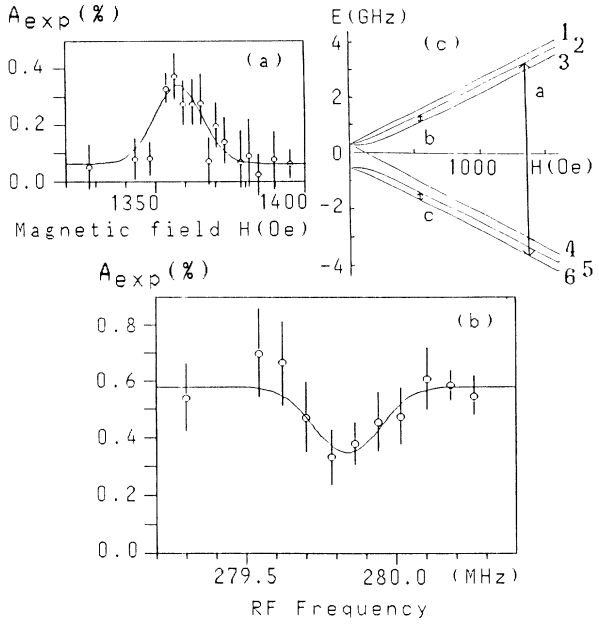

FIG. 3. The observed β-ray radiation-detected magnetic resonances. (a) A magnetic resonance observed with the transition a as a function of magnetic field at an applied microwave frequency of 6.891 GHz. (b) Magnetic resonances observed with the transitions b and c as a function of the applied rf frequency at 527 Oe. Solid lines in (a) and (b) are the results of a least χ-squares fit to the experimental data with a resonant Gaussian function plus quadratic background, where the position, height, and width of the peak and quadratic background were free variables to be determined. The error bars shown in (a) and (b) are statistical only. (c) The relevant transitions in the energy levels of divalent ^{170}Tm in SrF_2.

the result of the observed resonance with varying magnetic field H is shown. The observed resonance indicates that the microwave transition enhances the polarization by about a factor of 6 compared to that from the optical pumping alone.

Magnetic resonances corresponding to the transitions between the sublevels 2 and 3 [transition b in Fig. 3(c)] and also 5 and 6 (transition c) were also measured by fixing the magnetic field at 527 Oe and by varying the rf frequency, in which a frequency modulation of 160 kHz was applied at 400 Hz.[13] The results are shown in Fig. 3(b).[14] From the values of the resonant rf frequency and the applied magnetic field, the hyperfine coupling constant A_{170} of divalent ^{170}Tm in SrF_2 was thus for the first time determined to be 589.38(30) MHz. The accuracy of this value is almost 1 order of magnitude better than those in free atoms obtained by other methods.[15–17] The error of the determined value A_{170} is mainly due to the uncertainty of the applied magnetic field strength (± 2 Oe at 527 Oe); this error can be reduced to less than 0.01% with a more stabilized and uniform magnetic field. Neglecting a possible hyperfine anomaly, the magnetic moment μ_{170} of ^{170}Tm was thus obtained to be $0.2476(16)\mu_N$ from the newly determined hfs constant A_{170} of ^{170}Tm [589.38(30) MHz] with the known value[18] of A, I, and μ of the stable ^{169}Tm by the following relation:

$$\mu_{170}=\mu_{169}(A_{170}I_{170})/(A_{169}I_{169}) .$$

The present results for the hfs constant and the magnetic moment are given in Table I together with previous data.[15–17] It should be emphasized that the error of the deduced magnetic moment of ^{170}Tm mainly comes from the error of the previous data of the magnetic moment for stable ^{169}Tm, not due to our present data of A_{170}. The obtained value of the magnetic moment of ^{170}Tm is in agreement with previous results within errors.

In conclusion, we have shown that significant nuclear polarization of unstable ^{170}Tm can be achieved and efficiently detected by β-ray radiation-detected optical pumping in solids; this experiment is the first to directly observe the nuclear polarization of paramagnetic ions due to optical pumping in solids, thus giving for the first time the hyperfine coupling constant of unstable ^{170}Tm in SrF_2 and also giving an accurate magnetic moment. The method demonstrated here for the achievement of significant nuclear polarization will be applicable to nuclei with a half-life longer than about 100 msec, which is a strong advantage over other methods such as adiabatic demagnetization and nuclear orientation at low temperature. This observation together with the rather simple experimental arrangement for this method thus suggest profound potentiality of the method for the study of unstable nuclei far from the stability line. Also it is to be noted that the degree of nuclear polarization (about 4% in the present case) can be expected to increase appreciably in the case of atom implantation because in that case we can use pure crystals as implantation media. Such crystals have only a small number of paramagnetic impurity centers, so that no significant deterioration of nuclear polarization due to cross relaxations will occur even in magnetic fields less than 1 kOe. Since the optical as well as magnetic properties of rare-earth and transition-metal atoms have been well investigated,[19] this method of optical pumping in solids is expected to be applicable to many elements. Radiation detected magnetic resonance with optical pumping in solids thus provides a new powerful method to measure the magnetic moment of unstable nuclei in the rare-earth and transition-metal regions.

TABLE I. Hyperfine coupling constant A of divalent ^{170}Tm in SrF_2 and magnetic moment of ^{170}Tm obtained in the present experiment together with previous data.

	A(MHz)	$\mu(\mu_N)$
Present	589.38(30)[a]	0.2476(16)
	589(20)[b]	0.247(8)
Ref. 15	200(3)[c]	0.2476(36)
Ref. 16	195(13)[c]	0.247(4)
Ref. 17	199.0(6)[c]	0.2464(17)

[a]For divalent ^{170}Tm in SrF_2, transitions b and c.
[b]For divalent ^{170}Tm in SrF_2; transition c.
[c]For free atoms.

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[12]In a simple picture of nuclear-spin memory, the nuclear polarization is predicted to be constant, independent of the applied magnetic field, in disagreement with the experimental results.

[13]The transitions *b* and *c* have different frequencies due to the effect of the nuclear Zeeman interaction, the *g* factor of which is somewhat modified from the true value due to an admixture of the excited state Γ_8 to the ground state Γ_7; J. M. Baker and B. Bleany, Proc. R. Soc. London Ser. A **245**, 156 (1958); R. G. Bessent and W. Hayes, *ibid.* **285**, 430 (1965). The width of the resonance is mainly due to the superhyperfine interactions between the Tm ions and the neighboring fluorine atoms and also due to the unresolved contribution of the two transitions *b* and *c*.

[14]The value of the experimental asymmetry A_{exp} is about a factor of 2 larger than that for transition *a* [Fig. 3(a)], since these data were taken more than six months after the data of Fig. 3(a) when the contribution of the contaminant β rays from ^{89}Sr (half-life is 50.6 d) became almost negligible.

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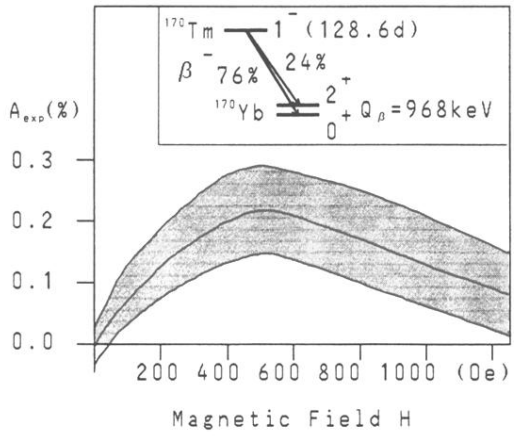

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